

Mantle refertilization and magmatism in old orogenic regions: The role of late-orogenic pyroxenites

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1	Mantle refertilization and magmatism in old orogenic
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27 Abstract

Pyroxenites and garnet pyroxenites are mantle heterogeneities characterized by a lower 28 29 solidus temperature than the enclosing peridotites; it follows that they are preferentially 30 involved during magma genesis. Constraining their origin, composition, and the interactions 31 they underwent during their subsequent evolution is therefore essential to discuss the sources of magmatism in a given area. Pyroxenites could represent either recycling of crustal rocks in 32 33 mantle domains or mantle originated rocks (formed either by olivine consuming melt-rock 34 reactions or by crystal fractionation). Petrological and geochemical (major and trace elements, 35 Sr-Nd and O isotopes) features of xenoliths from various occurrences (French Massif-Central, Jordan, Morocco and Cameroon) show that these samples represent cumulates crystallized 36 37 during melt percolation at mantle conditions. They formed in mantle domains at pressures of 38 1-2 GPa during post-collisional magmatism (possibly Hercynian for the French Massif-39 Central, and Panafrican for Morocco, Jordan and Cameroon). The thermal re-equilibration of 40 lithospheric domains, typical of the late orogenic exhumation stages, is also recorded by the 41 samples. Most of the samples display a metasomatic overprint that may be either inherited or 42 likely linked to the recent volcanic activity that occurred in the investigated regions.

The crystallization of pyroxenites during late orogenic events has implications for the subsequent evolution of the mantle domains. The presence of large amounts of mantle pyroxenites in old orogenic regions indeed imparts peculiar physical and chemical characteristics to these domains. Among others, the global solidus temperature of the whole lithospheric domain will be lowered; in turn, this implies that old orogenic regions are refertilized zones where magmatic activity would be enhanced.

49

50 **1. Introduction**

51 The shallowest part of the Earth's upper mantle is made primarily of peridotite, with spinel 52 lherzolite and harzburgite being the most abundant lithologies. Pyroxenites and eclogites 53 represent less than two percent of the upper mantle according to estimations based on isotopic 54 considerations (Pertermann and Hirschmann, 2003), or up to five percent considering estimations made in orogenic peridotite massifs (e.g., Kornprobst, 1969; Pearson et al., 1993). 55 Although they represent only a small volume of the upper mantle, pyroxenites and eclogites 56 57 provide important information about dynamic processes occurring since the formation of the 58 Earth, and about crustal recycling and melt circulation through the mantle (e.g., Downes, 59 2007; Gonzaga et al., 2010a). Due to their fertile composition and low solidus temperature compared to the host mantle peridotites, pyroxenites are quite relevant during mantle partial 60 61 melting and basaltic magma genesis (Hirschmann and Stolper, 1996; Lambart et al., 2009). 62 Their role in basalt genesis has indeed been inferred in oceanic and continental settings (e.g., Sobolev et al., 2005; Heinonen et al., 2013). 63

64 The origin of pyroxenites (as well as eclogites) found in mantle bodies or in mantle xenoliths 65 is highly debated. They can represent dehydrated or residual and partially molten crustal 66 lithologies recycled into the upper mantle through subduction zones (e.g., Allègre and Turcotte, 1986; Viljoen et al., 2005; Gonzaga et al., 2010a; Montanini et al., 2012). Such 67 68 mantle heterogeneities usually display a greater variability in chemical and isotopic 69 compositions compared to peridotitic mantle (e.g., Gonzaga et al., 2010a, b). Pyroxenites can 70 also represent high-pressure cumulates fractionated from basaltic magmas circulating through 71 the upper mantle (O'Hara and Yoder, 1967; Viljoen et al., 2005; Downes, 2007; Gonzaga et al., 2010a; Perinelli et al., 2011), creating a veined mantle below ancient magmatic provinces. 72 Alternatively, this veined mantle has been interpreted as representing a lithospheric 73

refertilization stage resulting from olivine-melt consuming and clinopyroxene-orthopyroxene
forming reactions (e.g., Bodinier et al., 2008).

76 The terminology in use (eclogite versus garnet pyroxenite) is mainly governed by the origin of 77 these rocks. In their review on eclogites and garnet pyroxenites, Gonzaga et al. (2010a) related 78 garnet pyroxenites to high-pressure mantle fractionation processes, and eclogites to recycling 79 of crustal protoliths. Historically, eclogites have been defined as rocks containing pyrope-80 garnet and omphacitic-clinopyroxenes (Haüy, 1822). Moreover, eclogites differ from garnet 81 pyroxenites in that they are more common in cratonic xenolith suites and have a higher jadeite component in clinopyroxenes (Pearson et al., 2005). Despite these differences, 82 83 multidisciplinary studies are still necessary to decipher the origin of a given sample, and 84 giving an *a priori* name is therefore hazardous. Hereafter, we will use the petrological term 85 garnet pyroxenite (garnet when more than 10% garnet is present, and pyroxenite when olivine 86 / [olivine+pyroxenes] < 40%, following the Streckeisen classification; Streckeisen, 1976).

87 The mineral assemblages developed in pyroxenites in response to changes in pressure and 88 temperature conditions are of great help in reconstructing the evolutionary stages they 89 underwent in the appropriate geodynamic setting (e.g., Montanini et al., 2006), whereas their 90 chemical and isotopic compositions can be used to decipher their origin and evaluate their 91 potential implication in magma genesis. The absence of modal olivine in mantle pyroxenites 92 is associated with the presence of garnet under spinel lherzolite stability conditions 93 (Kornprobst, 1969); this leads to the existence of two subfacies, namely the Seiland at lower 94 and the Ariegite at higher pressure conditions (O'Hara, 1967). In these two subfacies, the Al-95 bearing phases consist of plagioclase and/or spinel, and spinel and/or garnet, respectively. 96 These two subfacies subdivide the spinel lherzolite domain and allow us to determine more 97 precisely the pressure-temperature-time (P-T-t) evolution of the lithosphere than with 98 peridotitic samples.

Garnet Pyroxenites in Old Orogenic Regions

99 We present here a petrological and geochemical study of 16 mantle garnet pyroxenite 100 xenoliths (from Morocco, Jordan, Cameroon and France) that represent the rare material 101 available for petro-geochemical studies. The aims of this study are: (i) to evaluate the 102 similarities and/or differences between pyroxenite samples from the different regions; (ii) to 103 constrain the origin of the studied pyroxenites; (iii) to constrain the P-T-t evolution of the 104 studied pyroxenites within the mantle; (iv) to evaluate the relations between the determined P-105 T-t paths and the geodynamic history of the regional lithosphere; and (v) to discuss the 106 influence of these mantle heterogeneities on the subsequent evolution of the lithosphere.

107

108 **2. Regional settings and sampling localities**

In order to compare the geodynamic evolution and chemical and isotopic composition of pyroxenites, we studied samples from four localities, each with different geodynamic histories: the French Massif Central (Europe), Morocco and Cameroon (Africa) and Jordan (on the Arabian plate; Fig. 1).

113 <u>French Massif Central (FMC)</u>: The Massif Central (Fig. 1a) represents one of the biggest parts 114 of the Variscan belt of Western Europe. Alkaline volcanism developed throughout Cenozoic 115 times, with the last manifestation occurring in the Chaîne des Puys $6,900 \pm 110$ yr. cal. as the 116 lake Pavin maar (Juvigné et al., 1996). The origin of this volcanism is still debated, and is 117 either attributed to the presence of a thermal anomaly related to a mantle plume (e.g., Granet 118 et al., 1995) or to asthenospheric upwelling related to the nearby Alpine orogeny (Merle and 119 Michon, 2001).

Mantle xenoliths are present in several locations of the FMC. Two distinct domains, probably inherited from Hercynian times, are recognized N and S of an E-W line located ~45°30'N (Lenoir et al., 2000). Both domains have been variously depleted through melting ~360 Ma ago, and have been enriched by metasomatic fluids (carbonatitic to the North, and rather related to silicate-melts to the South; Wittig et al., 2007) likely related to the Cenozoic plume
(Zangana et al., 1997; Wittig et al., 2007).

126 Three of the studied samples come from the Devès volcanic field (S domain; LN-78 and LP-127 27 from Le Marais de Limagne, and SD-53 from Saint-Didier d'Allier), a basaltic plateau 128 emplaced between 2.7 and 0.6 Ma (Nehlig et al., 2001). Teleseismic tomography studies 129 indicate that the thermal anomaly in the mantle between 100 and 140 km below the Devès 130 area is the largest of the FMC (Sobolev et al., 1996). One sample comes from Le Pouget, near 131 Montpellier; this locality does not belong to the FMC but, instead, to the 'pyreneo-corso-132 sarde' Alpine belt. As the basement of this orogenic segment is also made of old Hercynian 133 lithosphere, the Le Pouget sample will be described together with the FMC samples. This last 134 sample has been briefly studied in previous works (Babkine et al., 1968; Fabriès et al., 1987).

<u>Cameroon (Fig. 1b)</u>: The oldest Cameroon terrains were structured during Eburnean times
some 2.1 Ga ago and were reworked during the Panafrican orogeny (Castaing et al., 1994).
Many granite bodies were emplaced around 520 Ma (Lassere et al., 1981).

138 Pyroxenites were sampled in the Youkou maar, in the Adamaoua volcanic plateau (Temdjim, 139 2006). This maar belongs to the Cameroon Volcanic Line (CVL; part of a still-active fault belt 140 in West Africa), which is characterized by important Tertiary and Quaternary alkaline 141 magmatism extending off-continent to several oceanic islands. Volcanism in this area started 142 around 60 Ma ago (Cantagrel et al., 1978) and is still active today. Mantle xenoliths (spinel 143 lherzolites being the most common) are present in volcanic rocks from both oceanic and 144 continental parts of the CVL. Several studies have emphasized a partial melting event 145 probably related to late Proterozoic crust formation (Lee et al., 1996). In many places, the 146 mantle rocks show evidence of interaction with enriched partial melts, probably related to the 147 Mesozoic breakup of Pangaea and the emplacement of the St. Helena mantle plume (Lee et 148 al., 1996; Caldeira and Munha, 2002; Temdjim et al., 2004).

149 Morocco (Fig. 1c): The middle Atlas area of Morocco is part of the Atlas Mountain range of 150 North Africa and was mainly structured during the Panafrican orogeny, lasting roughly from 151 700 to 530 Ma (Gasquet et al., 2005). The orogenic period was followed by the formation of 152 large Paleozoic sedimentary basins that were strongly folded during late Carboniferous-153 Permian compression (Pique and Michard, 1989). Cenozoic volcanism started before 35 Ma; 154 the last eruptions are probably not older than 0.5 Ma (El Azzouzi et al., 2010) and are 155 tentatively linked to a hot line spreading from the Siroua to Oujda (e.g., El Azzouzi et al., 156 2010).

Mantle xenoliths are present in volcanic rocks from the middle Atlas, and show a wide range of lithological and chemical heterogeneity. As in Cameroon, an older melting event is recorded by the chemistry of clinopyroxene (Raffone et al., 2009; Wittig et al., 2010). In most samples, this event has been overprinted by a widespread modal metasomatism (Pezzali et al., 2015). This metasomatic episode is probably very young (less than 200 Ma) and involves the percolation of alkaline melts with HIMU affinity as well as carbonatitic fluids (Raffone et al., 2009; Natali et al., 2013; Pezzali et al., 2015).

164 The Morocco samples come from the Bou-Ibalratene basaltic maar belonging to a volcanic 165 group of nearly 100 monogenic edifices oriented about N170°E, and located South of Azrou 166 in the middle Atlas.

Jordan (Fig. 1d): Jordan is located in the northern part of the Arabian-Nubian shield and was
affected by important metamorphism and magmatism during the Panafrican orogeny from 640
to 540 Ma ago (Stein and Goldstein, 1996). Large alkaline volcanic fields named 'Harrats' are
present all along the Arabian plate from Yemen in the South to Jordan and Syria in the North.
They are related to the Red Sea and Jordan rifts and were emplaced during Tertiary and
Quaternary times (Bertrand et al., 2003).

The Jordan samples come from three different maars belonging to the large, NW-SE trending, Harrat Ash Shaam that extends from SW Syria to the Northern part of Saudi Arabia. Studies concerning Harrat Ash Shaam mantle xenoliths have shown that the mantle in this region was affected by partial melting and refertilization events between 870 and 620 Ma ago. These processes are related to the formation of juvenile crust in a continental arc system (Krienitz and Haase, 2011).

179

180 **3. Analytical techniques**

181 This study is based on major and trace element and isotope analyses of whole rocks and 182 mineral separates (separated under binocular microscope for their purity after crushing, 183 sieving and washing). In-situ chemical analyses have been made on polished thin sections (30, 184 100 or 150 μ m thick). Details on analytical techniques and settings are given in 185 supplementary material.

186

187 **4. Petrology**

188 **4.1. Petrography**

189 The sample labels, rock types and modes of the studied samples, and the abbreviations used190 for minerals are presented in Table 1.

Pyroxenites from the FMC have recrystallized granoblastic textures with average grain size of 1mm for the samples from the Devès volcanic province (LN-78, LP-27 and SD-53); grain size is smaller in the sample from Le Pouget (pyroxenes ~0.25 mm, Spl and Grt ~0.75 mm). The prevailing mineral assemblage in all the samples is Cpx+Grt+Opx+Spl±Am. Grt is often located around green Spl and contains green Spl, Pl, Opx and Cpx inclusions that are surrounded by radial fractures (Fig. 2a-b). Those inclusions are remnants of a former paragenesis. In LN-78 larger Cpx grains (>2 mm) display Opx exsolutions (Fig. 2c). This sample is composite with an Am-free part and an Am-rich zone. In the three Devès samples, small grains (50 – 100 μ m) of Opx, Pl and brownish Spl are locally observed at the contact between Grt and Cpx (Fig. 2d). In all samples, Grt is surrounded by a thin (5 to 40 μ m; Fig. 2b, d) brownish kelyphite rim made of Opx-Pl-Spl (proportions 64:23:13%, respectively, estimated using mineral compositions and the method of France and Nicollet, 2010).

203 Samples TAK-3 and TAK-4 from Morocco have a granular texture with an average grain size 204 of 3 mm and show a penetrative foliation. The paragenesis is Grt+Opx+Cpx+Spl for TAK-3 205 and Grt+Cpx+Spl+Spr+Opx for TAK-4. In TAK-3, Grt occurs around the Spl demonstrating 206 an earlier crystallization of Spl, whereas in TAK-4, the Grt-Spl association is observed only 207 locally (Fig. 2e-f). In TAK-3 large Cpx grains (6 mm) contain Grt and Opx exsolutions, and 208 some very thin (1-2 µm) Spl exsolutions are observed in other Cpx. In TAK-4, Spr is present 209 as inclusions in Spl or Cpx, surrounding the Spl at the Grt contact (Fig. 2e-f), or as thin 210 exsolutions in Cpx. In this same sample a peculiar inclusion is observed in a Cpx grain, 211 containing a Spl+Spr+Pl+Opx assemblage (Fig. 2g). Both samples suffered extended Grt 212 kelyphitization (~0.4 mm large in average).

213 Petrographically, the Cameroon samples can be divided in two groups. The first group (YK-214 01, YK-05 and YK-16) represents initial Cpx megacrysts (0.5-3cm) that exsolve Grt grains up 215 to 500 µm wide (Fig. 2h-j). In YK-01 Cpx encloses Spl grains that are surrounded by 216 abundant Grt (Fig. 2h). YK-05 megacrysts record a deformation stage and exsolve Opx (Fig. 217 2i-j). The second group (YK-03, YK-12, YK-13) is characterized by a polycrystalline 218 Cpx+Opx+Grt+Spl assemblage (average grain size 3 mm). In these samples, Grt is either 219 exsolved from Cpx, or localized around Cpx grains and concentrated at the Spl rim. In YK-13, 220 Opx is also localized around Cpx grains and is probably exsolved. In this sample, some 221 apatite is observed, locally included in Grt or Opx. Thin Am exsolutions (~20 µm wide) are 222 also observed in some Cpx grains. In Cpx from all Cameroon samples, very thin (1-2 µm wide) Spl exsolutions are present; in samples YK-13 and YK-16, these can reach tens of μ m and are locally surrounded by Grt exsolutions. Am is present in all the samples except YK-01. A thin ($\leq 20 \ \mu$ m) kelyphitization zone is present around Grt, and a melting zone (< 15 μ m) is often observed at the contact between kelyphite and surrounding minerals. Later surface processes are documented by some carbonate veins that crosscut Grt kelyphite and melting zones.

229 The Jordan samples include Spl Grt pyroxenites (JO-7b and JO-7h) and Grt free 230 orthopyroxenites (JO-10e and JO-12h); JO-12h is an Am-bearing olivine orthopyroxenite. 231 The average grain size in all samples is 1 mm (Fig. 2k). In JO-7b and JO-7h, Grt (~50 µm) is 232 exsolved from pyroxene, around which it concentrates; some Grt also appears around green 233 (in JO-7b, Fig. 2k) or brown (in JO-7h) Spl. In JO-7h some very thin (~2 µm) Spl exsolutions are observed in Cpx. In JO-10e orthopyroxenite, rutile is observed along two exsolution 234 235 directions in Opx (Fig. 21); moreover, this sample is diffusely permeated by dikelets and 236 pockets of carbonate. In JO-7h, Opx is fractured and oxidized at its rim. In both JO-7b and 237 JO-7h, Grt is kelyphitized (~40 µm wide) at the margins.

238

4.2. Major element mineral chemistry

240 Mineral compositions are very homogeneous in each studied sample, allowing us to use 241 average compositions to compare the different samples (Table 2). Cpx and Opx are diopside 242 and enstatite, respectively. Their compositions are broadly similar in the samples from the 243 different provenances (Fig. 3a). In Cpx, wollastonite content is higher than 43%, ferrosilite 244 content lower that 10%, and jadeite content varies from 4.7 to 12.5%, whereas in Opx 245 wollastonite content is lower than 2%, ferrosilite content varies between 9 and 18%, and jadeite content is below 1%. TAK-4 Cpx are not stoichiometric; they are silica 246 247 (Si/(Ca+Na+Mg+Fe)=1.04) and Ca $(X_{Wo}=0.51)$ enriched. Grt are pyrope (Fig. 3b), with 248 compositions between 62 and 73% for the pyrope component (Mg) and between 11 and 24% 249 for the almandine component (Fe) at a nearly constant grossular (Ca) content (~13%). Spl show a wide range of compositions. The Cr₂O₃ vs. Al₂O₃ anti-correlation (R²=0.96) is related 250 251 to the Al-Cr substitution in Spl and shows a variation from 0 to 30% of the chromite component. For all the studied samples, the ratio Mg-component/ Fe^{2+} -component (spinel 252 253 s.s./hercynite) is around 2, and #Cr varies from 1 to 12% in all samples except JO-12h, for 254 which the value is 30.5% (with #Cr=Cr/(Cr+Al) in molar proportions). For the samples 255 displaying two Spl generations (LN-78, LP-27 and SD-53), the brown ones (second 256 generation) are poorer in chromite component than the primary green ones. Pl compositions 257 are variable, ranging from andesine in FMC samples (with anorthite content varying from 35 258 to 47 mol%), to bytownite in TAK-4 (Morocco; An₈₄). Analyzed amphiboles are 259 magnesiohastingsites in LN-78, YK-05 and YK-13, pargasites in YK-03 and ferro-edenite in 260 JO-12h. Olivine is Fo₈₉ in JO-12h.

261

262 **5. Mineral trace element compositions**

263 Trace element compositions of minerals have been obtained for Cpx and Grt in all samples, 264 and for Am, Pl, and Opx when these minerals were large enough (Table 3). Cpx trace element 265 composition is variable in the different samples, and sometimes within a single sample. In 266 FMC samples (Fig. 4a), the Cpx show variable light-rare earth elements (LREE) contents, 267 ranging from strongly depleted (in Le Pouget sample) to slightly enriched compared to the 268 medium-REE (MREE). They are also depleted in heavy-REE (HREE), reflecting their 269 chemical equilibrium with Grt (see section 8.1.2 "Cpx/Opx and Cpx/Grt partition 270 coefficients"). In sample LN78, some Cpx located close to Am grains are highly enriched in 271 LREE, less HREE depleted than other Cpx, and have REE content similar to associated Am 272 grains. In the two Morocco samples (Fig. 4a), the Cpx have REE normalized contents that 273 decrease from the LREE to the HREE region. In TAK-4 the decrease from the LREE to the 274 HREE is more pronounced than in TAK-3 and slight positive Eu and Sr anomalies are present 275 (Fig. 4b). Similar to their analogues from FMC, Cpx in the Cameroon samples are all HREE 276 depleted (Fig. 4c), in agreement with the presence of Grt. Their LREE composition is variable 277 and ranges from 0.6 to more than 10 times the chondrite value. Grt-bearing samples from 278 Jordan show two types of Cpx compositions (Fig. 4c); Cpx grains in contact with Grt are 279 strongly depleted in HREE relative to MREE, whereas those far from Grt are only slightly 280 depleted. Their LREE content is also variable, from largely depleted in JO-7b to slightly 281 enriched in JO-7h. In Grt-free samples, Cpx possess higher REE abundances and are LREE 282 enriched. In all these samples, Cpx have very low Nb, Ta and often Zr and Hf contents (Fig. 283 4b, d). All the analyzed Grt grains show typical REE patterns, highly enriched in HREE (up to 284 100x chondrite in TAK-3 from Morocco) in comparison to largely depleted LREE contents 285 (Fig. 4e). Some Grt display relatively flat patterns from MREE to HREE, reflecting the low 286 HREE contents of the whole rock. Grt is strongly enriched in U (Fig. 4f). Pl in FMC samples 287 is LREE enriched and shows strong Eu, Sr and Ba positive anomalies (Table 3).

288

289 6. Whole rock major and trace element compositions

290 Whole rock major element compositions are summarized in Table 4 and Figure 5. Mg# 291 (where Mg# = Mg/(Mg+Fe) in molar proportions) varies from 79.8 in SD-54 (FMC) to 89.3 292 in JO-7h (Jordan; Table 4). SiO₂ content ranges from 42.6 to 54.7 wt%; the lowest values are 293 found in the two samples from Morocco which are thus classified as ultramafic rocks while all 294 other samples are mafic rocks (Le Bas and Streckeisen, 1991). MgO content varies from 15.5 295 wt% in LP-27 (FMC) to 33.1 wt% in JO-12h (Jordan). Al₂O₃ content ranges from 3.2 wt% in 296 JO-10e (Jordan) to 22.5 wt% in TAK-4 (Morocco). CaO content varies from 1.7 wt% in JO-297 12h (Jordan) to 16.4 wt% in Le Pouget sample (FMC). FeO_t (all iron expressed as FeO)

298 content varies from 4.6 wt% in JO-7h (Jordan) to 8.1 wt% in JO-10e (Jordan). Na₂O content 299 ranges from 0.04 wt% in JO-12h (Jordan) to 1.41 wt% in YK-13 (Cameroon). In all the oxide-300 oxide composition plots, whole rock compositions plot between the main bearing minerals, 301 namely Cpx, Opx, Spl, and Grt. Some samples, especially those from Morocco, are 302 furthermore slightly shifted toward the Spl and/or Spr composition. MgO is correlated with 303 SiO₂, and anticorrelated with CaO and Al₂O₃. The average composition of primitive MORB 304 and alkaline melts are added in Figure 5, and are considered as possible proxies of a 305 prospective trapped melt in mantle domains.

306 Whole rock trace element concentrations are summarized in Table 4 and Figure 6. Whole rock 307 REE content for FMC pyroxenites show typical spoon-shaped patterns with high HREE 308 content, depletion in MREE and enrichment in LREE (Fig. 6a). The two different parts of LN-309 78 have parallel REE patterns, the Am-rich part being more LREE enriched; Le Pouget 310 sample possesses similar MREE and HREE composition, but is largely depleted in LREE, 311 with La lower than 0.1x chondrite, in agreement with its strongly LREE depleted Cpx (Fig. 312 4a). The two samples from Morocco have contrasting REE compositions; TAK-3 pyroxenite 313 shows a pattern similar to the FMC pyroxenites, whereas the Spr-bearing TAK-4 sample has a 314 very low HREE content (< 3x chondrite) coupled with a marked LREE enrichment (La > 35x 315 chondrite), and a small positive Eu anomaly associated with a positive Sr anomaly. Similar to 316 the FMC samples, two types of REE patterns are displayed by the Cameroon pyroxenites (Fig. 317 6c); they all have low HREE content, and most of the samples are LREE depleted. Only two 318 samples (YK-13 and YK-16) show LREE enrichment. The pyroxenites from Jordan also have 319 contrasted REE patterns (Fig. 6e). Three samples are LREE enriched, with (La/Yb)_n values 320 ranging from 2.5 to 7 and quite variable HREE compositions, while sample JO-7b is LREE 321 depleted.

In trace element spider diagrams (Fig. 6b, d, f), all samples present large negative Rb and Nb anomalies, associated with negative Zr anomalies (except for TAK-3 that displays a slight positive anomaly). Most of the samples also display a positive Sr anomaly; only JO-10e (Jordan) displays a negative Sr anomaly.

326

327 **7. Isotopic data**

Sr-Nd isotope data have been obtained on separated Cpx from four samples from Jordan, three from Cameroon, and two from Morocco (Table 5; not enough material was available for other samples). ⁸⁷Sr/⁸⁶Sr values are homogeneous and range from 0.702661 to 0.703375 (Fig. 7); in contrast, ¹⁴³Nd/¹⁴⁴Nd values are more variable and range between 0.512774 and 0.513433. Both the lowest and highest values are observed in Jordan samples. All the samples, except JO-7b and TAK-3, plot below the mantle array; most values are close to the HIMU component (Zindler et Hart, 1986).

335 50 analyses of oxygen isotopic ratios have been obtained on Cpx, Opx, Grt and Spl separates (Table 5). Most of the values fall within the mantle range for peridotites (Ionov et al., 1994; 336 Mattey et al., 1994a; Chazot et al., 1997; Zhang et al., 2000), with δ^{18} O ranging from 4.82 to 337 5.72‰ for Cpx, from 4.94 to 6.12‰ for Opx, from 5.15 to 5.97‰ for Grt, and from 4.33 to 338 339 4.97‰ for Spl (Fig. 8). Fractionation between Grt and Cpx is positive and close to 0.3‰ with 340 the exception of three samples from Cameroon having slightly negative fractionation values 341 (Fig. S1a). Fractionation between Opx and Cpx is also positive (except for two samples, Fig. 342 S1b), and in the range of values observed in mantle lherzolites from Yemen (Chazot et al., 1997). 343

344

345 **8. Discussion**

346 The various petrographical and geochemical data presented are discussed hereafter to 347 determine the last equilibration stage conditions (T, fO₂, section 8.1) of the studied Grt 348 pyroxenites, and to track their origin (recycled or not?, section 8.2) and the metasomatic 349 reactions they subsequently suffered (section 8.3). Pyroxenite pressure-temperature variations 350 through subsolidus evolution are reconstructed for each sample (section 8.4), and the apparent 351 inter- and intra-region variability (petrographical and geochemical) are discussed in section 352 8.5. Finally we present our hypothesis in a geodynamic framework to discuss implications in 353 terms of mantle refertilization (section 8.6).

354

355 **8.1. Last equilibration stage conditions**

8.1.1. Thermometry and oxybarometry

Equilibrium temperature (T) estimates are presented in Table 1. Opx-Cpx equilibrium T of 357 358 Grt-bearing peridotites and pyroxenites are best estimated using the Taylor (1998) 359 geothermometer (Wu and Zhao, 2012), and are as follows: 730°C and 930-1080°C (Le Pouget 360 and other FMC samples, respectively), 910°C (TAK-3, Morocco), 780-810°C (Jordan), and 361 900-980°C (Cameroon). Equilibrium T cannot be estimated for TAK-4 since it contains 362 aluminous diopside. For comparison, Opx-Grt equilibrium T, calculated using the Nimis and Grütter (2010) geothermometer, are as follows: 715-1020°C (FMC), 890°C (TAK-3, 363 364 Morocco), 720-760°C (Jordan), and 870-980°C (Cameroon). In general, equilibrium T 365 calculated with both geothermometers are consistent. Because 2+ cations diffuse faster than 366 3+ cations, T calculated using major element based thermometers are likely to be indicative of late stage equilibration T, whereas REE-in-two-pyroxene thermometers are expected to be 367 indicative of sub-magmatic T (e.g., Liang et al., 2013). Valid results have been obtained using 368 the Liang et al. (2013) model for two samples from the present study: LN-78: 1376 ±126 °C, 369

and LP-27: 1194 ± 69 °C; those equilibrium T are consistent with the record of an early magmatic stage.

Redox conditions of mantle domains are usually estimated using oxybarometers that 372 necessitate olivine bearing-rocks (e.g., Goncharov and Ionov, 2012), and are thus not 373 appropriate for the olivine-free pyroxenites studied here. Fe³⁺ concentrations in Cpx, Opx, and 374 375 Grt have been estimated using stoichiometric criteria following the classical approach of **Droop** (1987); corresponding $Fe^{3+}/\Sigma Fe$ values are reported in Table S1 and Fig. 9. Although 376 377 such calculation results should be taken with caution, good correlations exist between $(Fe^{3+}/\Sigma Fe)_{Opx}$ and $(Fe^{3+}/\Sigma Fe)_{Cpx}$ (R²=0.69), between $(Fe^{3+}/\Sigma Fe)_{Grt}$ and $(Fe^{3+}/\Sigma Fe)_{Opx}$ 378 $(R^2=0.85)$, and between $(Fe^{3+}/\Sigma Fe)_{Grt}$ and $(Fe^{3+}/\Sigma Fe)_{Cpx}$ $(R^2=0.71;$ Fig. 9). Those good 379 380 correlations strongly support the validity of the stoichiometric calculations for the present study: $(Fe^{3+}/\Sigma Fe)_{Cpx}$ ranges from 0 to 0.6, $(Fe^{3+}/\Sigma Fe)_{Opx}$ from 0 to 0.22, and $(Fe^{3+}/\Sigma Fe)_{Grt}$ from 381 0 to 0.21. $(Fe^{3+}/\Sigma Fe)_{Melt}$ was calculated using $(Fe^{3+}/\Sigma Fe)_{Cpx}$ values and the approach 382 383 formulated in France et al. (2010). Rough estimates for ΔFMQ (Fayalite-Magnetite-Quartz oxygen fugacity buffer) were obtained using the correlation between ΔFMQ and $(Fe^{3+}/\Sigma Fe)_{Grt}$ 384 presented by Goncharov and Ionov (2012). (Fe³⁺/ Σ Fe)_{Melt} is plotted against Δ FMQ in Fig. 9. 385

More generally, the FMC samples appear to be the most reduced pyroxenites with (Fe³⁺/ Σ Fe)_{Melt} ranging from 0.4 to 0.6; Jordan pyroxenites are slightly less reduced with an average (Fe³⁺/ Σ Fe)_{Melt} of 0.6; the average (Fe³⁺/ Σ Fe)_{Melt} for Morocco samples is 0.66; and Cameroon samples are the most oxidized samples studied herein with (Fe³⁺/ Σ Fe)_{Melt} ranging from 0.56 to 0.83 for an average value of 0.73 (Fig. 9).

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392 8.1.2. Cpx/Opx and Cpx/Grt partition coefficients

393 The major element compositional homogeneity of all the minerals among the studied samples, 394 and the presence in most of the samples of well-developed exsolution lamellae indicate that 395 (at least partial) intra-sample equilibrium was achieved; this is supported by the typical mantle 396 T calculated from the chemical composition of Cpx, Opx and Grt (Table 1). Oxygen isotope 397 data also show that no large disequilibrium exists between minerals in the studied pyroxenites. 398 This chemical equilibrium among minerals was also tested by trace element partition 399 coefficient calculations between Cpx and Opx, and between Cpx and Grt in the different 400 samples (Fig. 10). The calculated $\mathbf{K}_{D(Cpx-Grt)}$ values are in the range of values published by 401 Viljoen et al. (2005) for group II eclogites from South Africa, interpreted as crystallization 402 products of small-volume melts in the lithospheric mantle. Our values are also very similar to 403 those obtained in a Cpx megacryst aggregate crystallized from a MORB type magma in the lithospheric mantle of China (Huang et al., 2007), and to K_{D(Cpx-Grt)} determined 404 experimentally at pressures varying from 2 to 3 GPa (Johnson, 1994). K_{D(Cpx-Grt)} are 405 406 dependent on the major element phase compositions (e.g., Harte and Kirkley, 1997); 407 calculations accounting for mineral compositions align closely with K_{D(Cpx-Grt)} values 408 obtained in the present study (Fig. 10). These results support that Cpx and Grt have attained 409 chemical equilibrium at mantle conditions in all the studied samples before their fast transport 410 to the surface. $K_{D(CDX-ODX)}$ values are compared to results obtained by Raffone et al. (2009) on 411 lherzolites and harzburgites from Morocco (Fig. 10). Although a strong variability is observed 412 in our data (due to large errors on the trace element concentrations of Opx that are strongly 413 depleted in trace elements), $K_{D(Cpx-Opx)}$ are in the range of mantle values determined by 414 Raffone et al. (2009).

415

416 **8.2. Pyroxenite origin as mantle cumulates**

417 It is still a matter of debate whether pyroxenite xenoliths brought to the surface by volcanic 418 processes represent metamorphically transformed crustal rocks (of either continental or 419 oceanic origin) or are the final products of melt circulation, crystallization and evolution in the 420 mantle. Here we use highly detailed petrological observations and major and trace element
421 compositions, coupled with stable and radiogenic isotopic compositions to decipher their
422 origin.

423

424 **8.2.1.** Whole rock compositions as a proxy of initial cumulative assemblages

425 Whole rock major element compositions define relatively good correlations for SiO₂, 426 CaO and Al₂O₃ versus MgO content (Fig. 5), and Al₂O₃ and SiO₂ are also well correlated. The 427 pyroxenites studied here show an increase in SiO₂ content correlated to an increase in MgO 428 (Fig. 11), which is in contrast to classic liquid lines of descent that highlight melt 429 differentiation processes (e.g., for MORB, calc-alkaline, or alkaline rocks) characterized by a 430 decrease in MgO when SiO₂ increases. High concentrations of highly compatible elements 431 such as Ni can be used to identify cumulative rocks, in which their concentration is by 432 definition higher than in the corresponding equilibrium melts. The Ni content of the studied 433 pyroxenites (~300-900 ppm) is much higher than most primitive MORBs (~300 ppm, and 434 decreasing with differentiation; Fig. 11), indicating that they cannot represent liquids, but 435 rather are cumulates (either s.s. cumulates or cumulates resulting from percolative fractional 436 crystallization). At this stage of discussion, their origin as crustal or mantle cumulates cannot be deciphered. 437

Since the studied pyroxenites represent cumulative rocks, their whole rock composition (major and trace elements) is indicative of the modal proportion of the initial cumulative assemblage; for example, the high Ni content attests to the presence of Spl among the cumulative minerals. We now present evidence supporting the assemblages that are reported in Table 1. The presence of magmatic Grt in four samples (FMC samples, and TAK-3 from Morocco; Fig. 6) is confirmed by the whole rock enrichment in HREE relative to MREE. The major element composition and Ni content of those samples also indicate that Cpx-Opx-Spl 445 were part of the initial cumulative assemblage (red dotted samples in Fig. 5). Among the 446 studied pyroxenites, 3 samples from Cameroon (YK-01, YK-03, YK-12) display small but 447 significant positive Eu anomalies (Fig. 6), classically indicating magmatic Pl. However, 448 similar anomalies have been reported in spinel-bearing peridotitic Cameroon mantle, and 449 attributed to regional metasomatism (e.g., Temdjim et al., 2004). This signature being 450 regional, and in the absence of other evidence, we follow their interpretation and infer that Pl 451 was not part of the initial cumulative assemblage in Cameroon samples. Only TAK-4 from 452 Morocco displays coupled positive Eu and Sr anomalies (Fig. 6) that have not been reported 453 for peridotitic samples from this area, and should be attributed to the presence of Pl in the 454 initial cumulative assemblage. In this sample, the major element and Ni concentrations point 455 to the additional presence of Cpx and Spl in the initial cumulative assemblage (Fig. 5). Major 456 elements and Ni concentrations of Cameroon samples demonstrate that Cpx-Sp (±Opx) were 457 part of the initial cumulative assemblage in all samples (Fig. 5), consistent with petrographic 458 observations. Petrographic relations also suggest that apatite was present as a magmatic 459 mineral in YK-13. According to petrography, major elements, and Ni contents, Opx was the 460 main cumulative mineral in JO-10e and JO-12h, together with Ol-Spl-Cpx in JO-12h, and 461 Spl-Cpx in JO-10e; two pyroxenes and Spl were present in the initial cumulative assemblages 462 of JO-7b, and 7h (Fig. 5, Table 1).

In addition to the cumulative mineral record, Downes (2007) invoked a trapped melt component to account for the whole rock major element compositions of orogenic pyroxenites interpreted to be mantle cumulates. In order to test this hypothesis, the compositions of relatively primitive mantle melts (represented by primitive MORBs and alkaline basalts) have been added to all graphs of Fig. 5. Only two samples (LP-27 from FMC, and YK-13 from Cameroon) seem to record a slight influence of trapped melt in their whole rock composition, as highlighted by their high Na₂O contents. This hypothesis is consistent with the LREE enrichment of those samples; in addition YK-13 is the only sample to contain early magmaticapatite, also consistent with a possible trapped melt component.

The initial cumulative assemblages determined here will be used in section 8.4 to determine the in which mantle domain crystallization occurred. Finally the major and trace element whole rock compositions of the studied cumulative pyroxenites are only representative of the initial cumulative assemblage (nature of the phases and modal composition) and are thus not indicative of the composition of the parental melts that thus cannot be determined.

477 An alternative hypothesis to the cumulative origin of the studied pyroxenites is that Grt-478 pyroxenites form as refractory residues after partial melting of eclogitic metagabbros (e.g., 479 Montanini et al., 2012). However, residual Grt-pyroxenites are expected to preserve the initial 480 Eu and Sr anomalies that are characteristic of eclogites with a recycled origin (Marchesi et al., 481 2013). In addition, we have identified that the initial cumulative assemblages are, depending 482 on the studied pyroxenites, either Cpx-Opx-Grt-Spl or Cpx-Opx-Spl (±Pl, ±Ol, ±Ap; Table 1), 483 which is not consistent with residual assemblages derived from either gabbroic protoliths 484 (Montanini et al., 2012; Marchesi et al., 2013) or Pl-free pyroxenites or eclogites (Kogiso et 485 al., 2003; Lambart et al., 2009). Such a model is therefore not valid for the pyroxenites 486 studied here.

487 TAK-3 and TAK-4 Grt-bearing pyroxenites from Morocco are slightly off the general trends 488 observed for major elements (Fig. 5). These samples are shifted toward Spl and/or Spr 489 compositions in most oxide composition diagrams, suggesting that higher amounts of Spl 490 and/or Spr were present as primary magmatic phases in these samples. The possibility that Spr 491 is a liquidus phase in mafic to intermediate magmas at high pressure conditions (from 1.1 to 3 492 GPa) has been highlighted by Liu and Presnall (2000). However, these authors pointed out 493 that Spr has rarely been described as a magmatic phase in natural rocks as it mostly occurs as 494 a metamorphic mineral. Textural, petrological and geochemical evidence presented here does

495 not clarify an igneous or metamorphic origin of Spr; this will be further discussed in section
496 8.4 '*P*-*T*-*t path reconstructions*'.

497 In summary, the studied pyroxenites represent cumulative rocks that initially crystallized Cpx, 498 Opx, and for some samples Grt and Spl (with trace apatite in one sample; Table 1); Pl was 499 part of the initial cumulative assemblage in only one sample (TAK-4). Such cumulative 500 assemblages are not expected in lower crustal (continental & oceanic) settings where Pl is 501 usually the dominant phase. Additionally, Px±Spl±Grt cumulates are expected at shallow 502 mantle depths according to experimental petrology constraints (e.g., Green & Ringwood, 503 1967), and are very commonly described among mantle xenoliths (e.g., Kaeser et al., 2009; 504 Perinelli et al., 2011) or in orogenic ultramafic massifs (e.g., Downes, 2007). The likely 505 mantle origin of the studied cumulates can be further tested using isotopic constraints.

506

507 **8.2.2.** Isotopic constraints support a mantle protolith

508 The oxygen isotopic composition of minerals is useful for constraining mantle versus recycled 509 origins of pyroxenites and eclogites (e.g., Downes, 2007; Gonzaga et al., 2010a). Cpx, Grt, 510 Opx, and Spl analyzed in this study have oxygen isotopic values typical of mantle minerals 511 (4.8-6.1‰, Fig. 8; e.g., Mattey et al., 1994a; Chazot et al., 1997), in contrast to more variable 512 compositions of Cpx and Grt in recycled eclogites (2-10%; e.g., Mattey et al., 1994b; 513 Gonzaga et al., 2010a) and bulk oceanic crust (3.7-13.6%; e.g., Gregory and Taylor, 1981). 514 Deep crustal portions of oceanic crust (primarily Pl-rich cumulates) may display typical 515 mantle values (Gao et al., 2006), but are distinct due to strong Eu anomalies not observed in 516 the pyroxenites studied herein. Major element compositions of the studied pyroxenites do not 517 overlap the composition of neither volcanic, nor plutonic oceanic crust (Fig. 11), also strongly 518 supporting that studied pyroxenites do not derive from recycled oceanic crust. It should be 519 noted here that similar cumulative rocks have been recognized at Moho depth in the Kohistan

520 overthickened arc crust (35-45 km; Burg, 2011). The studied pyroxenites are therefore most 521 likely mantle cumulates; only TAK-4, which displays mantle like δ^{18} O values and magmatic 522 Pl, may represent either a shallow mantle or deep crustal cumulate. Inter-mineral δ^{18} O 523 variations are presented in supplementary figure S1; as a whole they are consistent with a 524 mantle origin.

525 Sr-Nd isotope data obtained on Cpx separates are used to test our previous conclusions. 526 Garrido et al. (2000) showed that ~65% of the Sr concentration of an ultramafic rock, and 527 ~15% of the Nd, is hosted in solid, melt, and fluid inclusions, as well as grain boundaries; Sr-528 Nd isotopic data obtained on Cpx separates (the main REE bearing mineral) should thus be 529 preferred to whole rock measurements. As a whole, the studied pyroxenites have Sr and Nd 530 isotopic ratios that are slightly above and below, respectively, the average DMM composition 531 (Fig. 7), in contrast to recycled eclogites that display more heterogeneous values than the host 532 mantle (Gonzaga et al., 2010a). Only two samples, TAK-3 and JO-7b (from Morocco and 533 Jordan, respectively), have higher Nd isotope ratios compared to DMM. The Nd isotopic 534 signature of JO-7b closely matches that of another garnet pyroxenite from Israel (BS-701 in 535 Stein et al., 1993) and peridotite samples from Saudi Arabia (Henjes-Kunst et al., 1990; 536 Blusztajn et al., 1995), and is probably a time integrated effect of a high Sm/Nd ratio (Sm/Nd 537 = 0.566; e.g., Borghini et al., 2013), which is the highest among all the samples. TAK-3 from 538 Morocco does not have a high Sm/Nd ratio, but its isotopic composition is very close to that 539 of lherzolite and websterite samples analyzed in the same area (Raffone et al., 2009). 540 Excluding TAK-3 and JO-7b, the Sr-Nd isotopic compositions of the studied pyroxenites, 541 although heterogeneous, overlap those obtained on mantle rocks from the various areas considered (Fig. 7), as is expected for pyroxenites with origins as segregations from mantle 542 melts (e.g., Downes, 2007; Gonzaga et al., 2010a). 543

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544 Our findings are in agreement with numerous previous studies identifying mantle pyroxenites 545 as mantle cumulates rather than the products of deep crustal recycling (O'Hara and Yoder, 546 1967; Frey, 1980; Downes, 2007; Griffin & O'Reilly, 2007; Perinelli et al., 2011). The 547 opposing view that mantle heterogeneities originate mainly from recycling associated with 548 subduction zones (Allègre and Turcotte, 1986) is not applicable to most of the studied 549 samples.

551 **8.3. Evidence for subsequent metasomatic interactions**

552 Most of the studied pyroxenites show evidence of late melt percolation and cryptic and/or 553 modal metasomatism. Two samples from Cameroon (YK-13 and YK-16) are LREE enriched 554 and sample YK-05, although LREE depleted, shows a slight enrichment in La compared to 555 Ce. These LREE enriched samples also have high concentrations in Ba, Sr, U and Th and low 556 concentrations in Zr and Hf. This metasomatic signature has already been observed in spinel 557 lherzolites from the Nyos volcano along the CVL (Temdjim et al., 2004), and is therefore a 558 regional overprint. The Morocco pyroxenites are also enriched in LREE, Ba, U, Th and Sr, 559 although more markedly in TAK-4 than in TAK-3. Raffone et al. (2009) documented similar 560 enrichments in peridotite xenoliths from the same volcanic province and showed that they are 561 related to alkaline melt percolation, probably associated with late Cretaceous or Eocene 562 volcanism. A metasomatic overprint is also present in three of the four studied Jordan 563 samples. Spidergrams highlight the LREE enrichment relative to MREE, and positive Ba, U, 564 and Th anomalies that are similar to metasomatic enrichments previously described in Jordan lithospheric mantle, and possibly related to Pan-African subduction (Shaw et al., 2007). 565 566 Metasomatism is also evident in the pyroxenites from the FMC. All the whole-rocks, except the southernmost sample (Le Pouget), have spoon-shaped REE patterns with decreasing REE 567 568 content from Lu to Nd, and are enriched in LREE (La, Ce and Pr), clearly pointing to LREE

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depleted protoliths, further enriched by LREE-rich percolating melts. These samples are alsohighly enriched in U and Th, and sometimes depleted in Nb, Zr and Hf.

571 LN-78 is a composite sample with an Am-bearing and an Am-free part. In the Am-bearing 572 part, Cpx is more LREE-enriched, and is in chemical equilibrium with coexisting Am as attested by the $D_{RFF}^{Cpx/Am} \approx 1$ (Chazot et al., 1996). In the Am-free part, Cpx has complex REE 573 patterns with HREE decreasing from Eu to Lu due to preferential incorporation of these 574 575 elements in coexisting Grt. Rare earths were initially decreasing from Eu to La (similar to Le 576 Pouget sample), but La and Ce have been selectively enriched during melt percolation and 577 crystallization of Am and Cpx in the Am-bearing part of the sample; similar conclusions have 578 been proposed by Downes et al. (2003). Similar metasomatic overprints with LREE, U, and 579 Th enrichments, and Zr-Hf and Nb-Ta depletions have been observed in peridotite xenoliths 580 from different localities in the FMC (Lenoir et al., 2000, Féménias et al., 2003, Dautria et al., 581 2010); those overprints are ascribed for most of the elements to Variscan (or late Variscan) --582 possibly subduction-related -- metasomatism (Lenoir et al., 2000; Féménias et al., 2003). 583 Other authors link this metasomatic event to the arrival of a mantle plume head in the early 584 Cenozoic (Dautria et al., 2010). Some xenoliths originated in the southern part of the FMC do 585 not show any metasomatic overprint (Dautria et al., 2010), in agreement with the lack of 586 enrichment in the Le Pouget sample.

A metasomatic overprint may be hard to distinguish from a trapped melt component present in the initial cumulative assemblage. Na content may be used as it is not expected to be particularly enriched by metasomatic fluid percolation in comparison to other major elements, however it is useful to track a trapped melt component in cumulates (see section 8.2.1, and Fig. 5). Among the studied samples only LP-27, and YK-13 have been shown to record a trapped melt component; all trace element characteristics discussed above for the other samples are likely related to metasomatic enrichments. As discussed in Pezzali et al. (2015), the samples displaying the weakest metasomatic overprint can be considered as geochemical remnants of the pre-metasomatic lithospheric mantle. Those are Le Pouget sample for FMC lithospheric mantle, Jo-7b for Jordan lithospheric mantle, YK-01, and YK-12 for Cameroon lithospheric mantle.

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599 **8.4. P-T-t** path reconstructions and pyroxenite evolution within mantle domains

Various geochemical data presented here have highlighted an origin as mantle fractionated melts for the studied samples. Precise petrological observations and whole rock trace element contents are here used to constrain the P evolution of these cumulates and to reconstruct the corresponding P-T-t paths. To proceed, the P-T stability fields, the associated facies and subfacies, and the corresponding subsolidus reactions separating lithospheric mantle domains, documented in Fig. 12a, will be considered. Initial cumulative assemblages, and the sequence of subfacies crossed during the P-T-t evolution are recalled in Table 1.

607 FMC pyroxenites display recrystallized textures resulting from various sub-solidus mineral 608 reactions. Grt entered the mineral assemblage as a liquidus phase (section 8.2), consistent with 609 a first crystallization of the FMC pyroxenites in the Ariegite subfacies (Fig. 12b). However 610 the numerous inclusions (green Spl, Cpx, Opx and Pl; Fig. 2a-b) observed in Grt show that the 611 present-day Ariegite-subfacies mineral assemblage (Cpx+Grt+Opx+Spl±Am) does not 612 represent that of the initial cumulate, but rather derivates from metamorphic reactions. This 613 mineral assemblage preserved as inclusions in Grt thus testifies for an early Seiland 614 pyroxenite facies assemblage. Some large Cpx grains (up to 2 mm) containing Opx 615 exsolutions (Fig. 2c) have escaped the general recrystallization process and are still 616 reminiscent of an early cooling. A demixing T of ~1275°C is estimated for sample LN-78 617 using image processing, mineral compositions, and Carlson and Lindsley (1988) relations. In 618 all the FMC samples but Le Pouget pyroxenite, Grt grains in contact with Cpx grains undergo

619 a later reaction leading to the local development of Pl, Opx and brown Spl (Fig. 12b), marking 620 the return of the samples into the Seiland P-T domain. P-T-t paths for FMC samples are 621 therefore summarized as follows (Fig. 12b): samples crystallized in Ariegite subfacies 622 conditions (magmatic Grt), suffered an early T decrease (large Cpx grains with Opx 623 exsolutions), and then underwent a concomitant P and T decrease that allowed a transition 624 towards the Seiland subfacies (testified by Opx-Spl-Pl inclusions in present-day Grt); a further 625 decrease in T brought the samples back into the Ariegite domain crystallizing the present-day 626 Grt that contains relics of the former Seiland stage (Opx-Spl-Pl in Grt). A final decompression 627 (or reheating) event that once again shifted some samples into the Seiland stability field.

628 A similar approach is followed to reconstruct the P-T-t evolution of the Morocco samples 629 (Fig. 12c). Grt was a liquidus phase in TAK-3 (Ariegite facies conditions; section 8.2) but was 630 absent when TAK-4 crystallized at a shallower depth. TAK-3 also contains some large Cpx 631 grains with Opx exsolutions, indicating a T decrease after crystallization. Grt currently present 632 in TAK-3 are mainly located around Spl (Fig. 2e-f) and formed by the reaction Opx+Spl+Pl-633 >Grt+Cpx that documents the transition from the Seiland to the Ariegite subfacies (a previous 634 transition from the Ariegite facies crystallization conditions to the Seiland subfacies is not 635 documented by the mineralogy). Although TAK-4 did not crystallize in Ariegite facies 636 conditions (no liquidus Grt, but magmatic Pl), abundant and large Grt and Cpx grains 637 associated with the absence of Ol provide evidence of subsolidus equilibration under those 638 conditions. The presence of a small Spr rim between Spl and Grt (Fig. 2e-f) suggests that the 639 transition from Seiland to Ariegite subfacies conditions was mainly governed by a decrease in 640 T inducing the reaction Opx+Spl+Pl->Grt+Spr; this is consistent with the Al-rich composition of TAK-4 Opx that is required for such a reaction (France et al., 2009). Alternatively, if TAK-641 642 4 was crystallized at deep crustal levels (see section 8.2.2), the cooling stage associated with 643 this reaction may have been associated with a pressure increase. We will hereafter consider

644 only the less complex model that necessitates only a cooling stage. The presence of numerous 645 Spr inclusions in Spl (Fig. 2e-f) and of one Spr relic surrounded by small crystals of Opx, Spl 646 and Pl within a Cpx grain (Fig. 2a) document a further reaction, namely Grt+Spr-647 >Opx+Spl+Pl, before the sample equilibrated in the Ariegite subfacies. This reaction marks 648 the transition from the Spr-bearing Ariegite subfacies to the Spr-absent Ariegite subfacies, 649 probably in response to a decrease in P. Despite those P-T-t estimates, Spr-bearing TAK-4 650 appears to have a rather complex paragenesis evolution. Further constraints from 651 thermodynamic models will be needed to decipher a more precise evolution, however such 652 work is beyond the scope of the present study. P-T-t paths for Morocco pyroxenites are 653 therefore mostly characterized by a cooling event subsequent to crystallization that it is 654 possibly followed by a concomitant P and T decrease before re-equilibration at lower T.

655 Pyroxenites from Cameroon contain large amounts of Cpx that exsolves Grt and sometimes 656 Opx. Grt was not a liquidus phase when these cumulates crystallized, as shown by their 657 relatively flat whole rock REE patterns (Fig. 6), constraining their formation in the Seiland 658 subfacies (Fig. 12b). As previously discussed for the FMC and Morocco samples, exsolution 659 of Opx is related to a decrease in T after crystallization. The occurrence of Grt is also related 660 to a strong cooling event when it exsolved from Cpx or crystallized around Spl while the 661 samples were moved to the Ariegite facies. This evolution can be accounted for by an isobaric 662 T decrease or a concomitant T and P decrease. P-T-t paths for Cameroon pyroxenites therefore record a rather simple P-T-t trajectory marked mainly by a decrease in T (Fig. 12b). 663 664 Evolution in pressure is poorly constrained, and a slight decompression may have occurred.

Pyroxenites from Jordan show only limited mineralogical reactions that can be used to decipher their P-T-t evolution (Fig. 12c). Pyroxenes from JO-12h are the most magnesian among the studied samples, Spl contains the highest amount of Cr_2O_3 , and ~10% Ol is present; altogether those observations point to a parental melt that was slightly more primitive 669 than for other samples. In this pyroxenite, the absence of Pl and the presence of 670 Ol+Cpx+Opx+Spl indicates that the reaction separating the Seiland subfacies and the Pl-671 lherzolite facies (Cpx+Opx+Spl->Ol+Pl) has not occurred. JO-12h therefore equilibrated in 672 the Seiland subfacies. Grt was not a liquidus phase in any of the Jordan samples (Fig. 6; 673 section 8.2), also implying crystallization in the Seiland subfacies. When present, Grt appears 674 as exsolutions in pyroxene or around Spl (JO-7b and JO-7h), and so is clearly a subsolidus 675 mineral. Grt exsolutions in pyroxenes are also indicative of a marked cooling event from the 676 Seiland to the Ariegite field. Also consistent with an important cooling event are the 677 numerous rutile exsolution lamellae in Opx of JO-10e (Fig. 21). Indeed during crystallization 678 at high pressure, in the absence of Ol and Spl, Ti is accommodated by Opx, and can be 679 exsolved to form rutile exsolution needles during cooling (Verhoogen, 1962; Moore, 1968). 680 P-T-t paths of Jordan samples are therefore mainly governed by a strong temperature decrease 681 but are not well constrained in pressure (Fig. 12c). As is the case for Cameroon samples, a 682 slight decrease in pressure may have occurred during thermal equilibration toward cooler 683 conditions (Fig. 12c).

684

685 **8.5. Inter- and intra-sites variability**

686 Invoking a common cumulative process for the formation of the studied pyroxenites may 687 sound odd given the inter- and intra-region petrographical and geochemical variability of the 688 samples. Nevertheless, partial crystallization of mantle derived melts during interactions with 689 peridotites at various depths has the potential to crystallize such apparently heterogeneous 690 cumulates (Downes, 2007). In particular, whole rock compositions (major and trace elements) 691 are only representative of the initial cumulative assemblage (\pm metasomatism) that has been 692 sampled by a given xenolith (Fig. 13). An extreme case would be that a small size xenolith 693 might sample only one mineral of this initial cumulative assemblage (e.g., Cpx, Opx, Grt, Spl;

694 cases 1, 2, 3 in Fig. 13). A more likely case would be that a xenolith samples a poly-mineral 695 piece of the initial cumulative assemblage (cases 4, 5 in Fig. 13). In that case, the whole rock 696 composition represents a mixing of the mineral compositions that relies on the modal 697 proportion within the initial cumulative assemblage (Fig. 13b-c). One striking feature is that 698 the modal composition of a given xenolith does not represent the modal proportion of the 699 initial cumulative assemblage as cumulates have suffered several metamorphic reactions 700 during their subsolidus evolution (Fig. 12, 13). The present day modal composition of a given sample is therefore not directly representative of the whole rock composition (e.g., TAK-4 701 702 contains 56 % Grt when the trace element content shows that Grt was not part of the initial 703 cumulative assemblage). The widespread metasomatic events that have affected most of the 704 samples are expected to modify mainly the trace element budgets (e.g., LREE enrichments; 705 samples 1, 2, 4, 5 in Fig. 13c).

Given the various crystallization pressures (Seiland versus Ariegites subfacies; Table 1), the numerous initial modal proportions that a given xenolith may sample (Fig. 13), and the range of metasomatic enrichments that can affect the samples (Fig. 13c, and section 8.3), it is likely that the samples display a strong inter- and intra-region variability, though they share a common genesis.

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712 **8.6. Geodynamic implications**

713 **8.6.1. Late-orogenic genesis of pyroxenites**

Petrological and geochemical evidences reported above, highlight that all the studied pyroxenite xenoliths are products of melt crystallization at mantle conditions (1-2 GPa). Similar cumulative rocks to those studied here have been recognized at Moho depth in overthickened arc crust (Burg, 2011) where they crystallized prior to the thinning and exhumation of the thickened crust. Cumulates have later undergone subsolidus evolution 719 under progressively changing P and T conditions; reconstructed P-T-t paths therefore also 720 highlight the P-T-t evolution of the regional host mantle, and can be used to discuss the 721 geodynamic settings that resulted in pyroxenite crystallization. Two P-T-t path types are 722 observed among the samples: the first is characterized by a strong cooling event that may have 723 been accompanied by a pressure decrease (Cameroon and Jordan samples; Fig. 12), and the 724 second is characterized by two important cooling stages separated by a period during which P 725 and T decrease jointly (FMC and Morocco samples; Fig. 12). The four studied areas have 726 been implicated in orogenic episodes in the last 800 Ma (either Panafrican or Hercynian). In 727 this perspective, the crystallization of mantle melts at depth, followed by a strong decrease in 728 T, and then by a decompression stage, can only occur during the exhumation of an orogenic 729 domain, and the associated thermal re-equilibration. Similar P-T paths for mantle rocks have 730 been described in different areas (Müntener, 1997; Fabriès et al., 1998; Kaeser et al., 2009), 731 and major magmatic episodes (e.g., magmatic underplating) are well known to occur in the 732 late evolution of orogenic domains (e.g., Henk et al., 1997; Fabriès et al., 1998; Cottin et al., 733 1998; Féménias et al., 2003; Kaeser et al., 2009; McCarthy and Müntener, 2015). Along with 734 underplating occurring at Moho depth, large circulations of melts in deeper mantle domains 735 are expected and associated with refertilization reactions (e.g., McCarthy and Müntener, 736 2015). The melting event may start during the first stage of exhumation when mantle is 737 upwelling roughly adiabatically (isostatic re-equilibration). Post-crystallization equilibration 738 to the local geothermal T is recorded by the first part of the cooling stage. Following 739 crystallization and cooling, Morocco and FMC (and possibly Cameroon and Jordan) samples 740 underwent decompression tentatively related to the continuation of erosion and tectonic 741 collapse of the orogenic edifices. All the studied cumulates record the late orogenic stage 742 corresponding to the thermal re-equilibration along the continental geotherm (last cooling 743 stage). FMC samples from the Devès volcanic field have recorded a late P decrease; this event could be linked to the Cenozoic bulging affecting the FMC (e.g., Sobolev et al., 1996;
Seranne, 1999). This is consistent with the absence of this last P drop in the southernmost
sample (Le Pouget), as no bulging has been described in the southern part of the FMC (e.g.,
Sobolev et al., 1996; Seranne, 1999).

748 The late-orogenic origin of the studied cumulates can be challenged by chronological 749 constraints; indeed Nd isotopic compositions can be used to put some constraints on the 750 timing of these events (e.g., Bizimis et al., 2005). Although these estimates should be 751 considered with serious caution (e.g., Wittig et al., 2006; Downes, 2007), building time 752 estimates using Nd in Cpx seems appropriate as those minerals host ~80% of the whole rock 753 Nd budget (Garrido et al., 2000). Assuming that in each region the pyroxenites formed from a 754 similar melt with homogeneous isotopic composition, samples from Cameroon have crystallized between 700 and 1000 Ma ago. Such an estimation is valid for Jordan 755 756 pyroxenites, for which the origin can be estimated between 550 and 650 Ma. Morocco 757 samples are strongly overprinted by metasomatic circulations, and although time estimates are 758 consistent with late Panafrican orogeny (~520 Ma), those samples will not be considered here. 759 For Cameroon and Jordan, these rough time estimates correspond, with large uncertainties, to 760 the main Panafrican orogenic events affecting the crust and the upper mantle in these areas. 761 Again, these time constraints should be considered with serious caution as most of the 762 samples have been affected by post-crystallization metasomatism, which may have modified 763 Sm/Nd ratios and Nd isotopic compositions.

As a whole, the pyroxenites were formed in mantle conditions during late-orogenic magmatism associated with orogenic exhumation (Fig. 14a-c). The recorded orogenic cycle is probably the last main one in each region, and may be Panafrican for Jordan, Morocco and Cameroon, and Hercynian for FMC. During their subsequent evolution, cumulates have suffered several interactions with percolating fluids and melts. 769

770 **8.6.2. Mantle refertilization and forthcoming magma genesis**

771 We have shown that the studied Grt pyroxenites are mantle cumulates that crystallized during 772 the relaxation following orogenic events, and subsequently underwent modal and cryptic 773 metasomatism. Pyroxenites are fertile lithologies as their solidus is crossed at lower T than the 774 lherzolite solidus; up to 15% pyroxenite partial melting can be reached before the onset of 775 lherzolite melting (Hirschmann and Stolper, 1996). The addition of a pyroxenite component in 776 a given mantle domain consequently corresponds to a refertilization stage (e.g., Bodinier et 777 al., 2008), and the presence of pyroxenitic and metasomatic lithologies in the mantle has 778 strong implications on basalt genesis (either MORB or OIB; Allègre and Turcotte, 1986; 779 Hirschmann and Stolper, 1996; Hirschmann et al., 2003; Kogiso et al., 2003; Kogiso et al., 780 2004; Pilet et al., 2008). The present study thus highlights that a refertilization stage of 781 lithospheric mantle can occur during the late orogenic re-equilibration period. It follows that 782 subsequent magmatic activity can be enhanced in old orogenic regions (such as FMC, 783 Morocco, Cameroon, Jordan), as it can be initiated earlier, and may be characterized by higher 784 magma volumes than in a non-affected region (Fig. 14). Nevertheless, tracking the implication 785 of such fertile lithologies in recent volcanism remains elusive as the melt-peridotite reactions 786 that occurred during melt ascent prevent the geochemical tracking of this influence (e.g., Le 787 Roux et al., 2009; Lambart et al., 2009).

788

789 **9.** Conclusions

The petrographical, geochemical, and isotopic study of garnet bearing pyroxenite xenoliths from four localities (FMC, Morocco, Jordan and Cameroon) allows characterization of their genesis and subsequent evolution. The whole-rock major element compositions and compatible element abundances, the absence of Pl in the initial cumulative assemblage for all

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794 the samples (but one sample from Morocco), and mineral oxygen isotopic compositions 795 highlight a cumulative mantle origin. Metamorphic reactions, exsolutions and trace element 796 whole-rock concentrations demonstrate that crystallization pressures ranged between 1 and 2 797 GPa (30-60 km depth) depending on the samples. P-T-t paths based on well characterized 798 petrographical relations and calculated equilibrium temperatures attest to important 799 movements in the respective lithospheres, whereas the presence of some hydrous minerals 800 such as amphiboles, and incompatible element concentrations attest to a metasomatic 801 overprint in most of the samples; this metasomatic enrichment also affects the host mantle. In 802 their geodynamical context, the samples are interpreted as representing the crystallization of 803 percolating melts formed at mantle conditions during exhumation of orogenic domains. 804 Although variable due to the long-term integration of various father/daughter ratios, the Sr 805 and Nd isotope signatures are consistent with crystallization during Panafrican times for 806 Cameroon, Morocco and Jordan samples.

807 The results presented here highlight that exhumation of orogenic domains and the associated 808 sub-adiabatic decompression trigger mantle partial melting; the melt percolating the mantle 809 crystallizes, at least partially, to form the studied cumulative pyroxenites. At mantle 810 conditions, pyroxenites are low melting point lithologies; their crystallization in mantle 811 domains therefore implies its refertilization. As a consequence, the magmatism in old 812 orogenic regions such as FMC, Morocco, Cameroon and Jordan, will be enhanced, and old 813 orogenic domains would be precursor zones for magmatism that would be initiated earlier and 814 would be volumetrically larger than in non-affected areas.

815

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- 1139

1140 *Captions:*

1141

Figure 1: Simplified maps and sampling localities (red stars) for FMC samples (a, modified after Wittig et al., 2007), Cameroon samples (b, modified after Temdjim et al., 2004),
Morocco samples (c, modified after Wittig et al., 2010), and Jordan samples (d, modified after 1145 Shaw et al., 2007).

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1147 Figure 2: Photomicrographs of the studied samples. a) Pl inclusion with radial fractures in the 1148 host garnet (FMC; sample LN-78; plane-polarized light); b) Green-Spl inclusion with radial 1149 fractures in host Grt; Grt grains are rimmed by a brown kelyphite displaying a Pl-Opx-Spl intergrowth (FMC; sample LN-78; plane-polarized light); c) Large Cpx porphyroclast with 1150 1151 Opx exsolutions (FMC; sample LN-78; cross-polarized light); d) Small Opx-brown-Spl-Pl 1152 association at the contact of Grt+Cpx. Green-Spl inclusions are observed in Grt. Grt are 1153 rimmed by a thin brown kelyphite displaying a Pl-Opx-Spl intergrowth (FMC; sample LN-1154 78; plane-polarized light); e-f) Association of Grt, Cpx, Spl, and Spr in the Morocco sample 1155 TAK-4. Grt is highly kelyphitized (brown). Spr rims the Spl grains included in Grt, and is 1156 present as inclusions in Spl grains (e: plane-polarized light; f: cross-polarized light). g) RGB 1157 image of a composite inclusion in a Cpx of the Morocco sample TAK-4; it is composed of 1158 Spr, Spl, Pl and Opx. Colors are: R=Si+Mg+Al, G=Si+K+Mg, B=K, (blue areas are altered 1159 and composed of zeolites); h) Modal image of sample YK-01 from Cameroon. Spl (blue) is 1160 rimed by Grt (pink). Grt is also exsolved from Cpx (green) megacrysts. Photomicrographs of 1161 thin section (i; cross-polarized light) and modal image (j) of YK-05 sample from Cameroon. 1162 Note the deformation of Cpx (green in (j)) grains that is highlighted by different polarization 1163 levels (i) and by the Opx (yellow in (j)) and Grt (pink in (j)) exsolutions. k) Sample JO-7b 1164 from Jordan; Cpx, Opx, green-Spl and Grt are observed, the Grt forms small white grains and

brownish kelyphite around Spl grains. I) Photomicrograph (cross-polarized light) of an Opx
grain in sample JO-10e from Jordan; the Opx is extinct and contains rutile exsolutions in two
crystallographic directions.

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Figure 3: Mineral chemistry. a) Cpx and Opx compositions in molar proportions (Wo: wollastonite component; En: enstatite component; Fs: ferrosilite component). b) Grt compositions in molar proportions (Alm: almandine; Spess: spessartine; Grs: grossular; And: andradite). Open squares: Cameroon samples; filled squares: Jordan samples; open circles: French Massif-Central samples; filled circles: Morocco.

1174

Figure 4: Chondrite-normalized (Anders and Grevesse, 1989) REE (**a**, **c**, **e**), and trace element (**b**, **d**, **f**) spider diagrams for Cpx, and Grt (Table 3). **a-b**) Cpx from French Massif-Central and Morocco samples, **c-d**) Cpx from Cameroon and Jordan samples, **e-f**) Grt for all samples.

1179

1180 Figure 5: Whole rock major element and Ni composition diagrams (Table 4). In all diagrams, 1181 mineral composition fields are added. The dashed lines highlight correlations. Red star 1182 represents the composition of primitive MORB taken as one composition of possible trapped 1183 melt (MORB composition is taken from Rubin and Sinton, 2007; an average of 3092 data 1184 points from slow spreading ridges is used as those melts are less affected by the various 1185 processes that may occur at crustal levels in fast spreading settings, e.g., France et al., 2014); composition (in wt %) is SiO₂=50.77; TiO₂=1.38; Al₂O₃=15.37; FeO_T=9.60; MgO=7.79; 1186 1187 CaO=11.71; Na₂O=2.45; K₂O=0.19; P₂O₅=0.15. Green star represents the composition of 1188 primitive alkaline basanite from Ethiopia taken as one composition of possible trapped melt 1189 (R. Pik personal communication, unpublished data); composition (in wt %) is $SiO_2=43.5$;

1190 TiO₂=2.6; Al₂O₃=12.5; FeO_T=11.0; MgO=10.4; CaO=11.9; Na₂O=2.5; K₂O=1.0; P₂O₅=0.5.

- 1191 Trapped melt seems to influence the Na₂O content for some samples.
- 1192

Figure 6: Chondrite-normalized (Anders and Grevesse, 1989) REE (a, c, e) and trace element
(b, d, f) spider diagrams for whole-rock pyroxenites. a-b) French Massif-Central and
Morocco samples, c-d) Cameroon samples, e-f) Jordan samples.

1196

Figure 7: ¹⁴³Nd/¹⁴⁴Nd *versus* ⁸⁷Sr/⁸⁶Sr for clinopyroxene separates from Morocco, Jordan, and 1197 Cameroon. Data for DMM (¹⁴³Nd/¹⁴⁴Nd=0.51313, ⁸⁷Sr/⁸⁶Sr=0.70263), and BSE 1198 $(^{143}Nd/^{144}Nd=0.512638, {}^{87}Sr/^{86}Sr=0.7045)$ are from Workman and Hart (2005), and for HIMU 1199 (¹⁴³Nd/¹⁴⁴Nd=0.51285, ⁸⁷Sr/⁸⁶Sr=0.70285) from Hart et al. (1992). Data are compared to 1200 regional mantle compositions (i.e., mantle xenoliths) in a), and to regional lava compositions 1201 1202 in b). Literature data: Mantle rocks (a) from Jordan (Henjes-Kunst et al., 1990; Stein et al., 1203 1993; Blusztajn et al., 1995; Baker et al., 1998), Morocco (Raffone et al., 2009), and 1204 Cameroon (Lee et al., 1996); and lava compositions (b) from Morocco (El Azzouzi et al., 1205 1999), Cameroon (Halliday et al., 1988; Lee et al., 1994; Ballentine et al., 1997; Marzoli et 1206 al., 2000; Rankenburg et al., 2005; Kamgang et al., 2008), Jordan (Shaw et al., 2003; Krienitz 1207 et al., 2009).

1208

Figure 8: Oxygen isotopic composition of the constituent minerals in the studied pyroxenites, δ^{18} O is expressed in ‰. Oxygen isotopic composition of corresponding minerals in peridotites are indicated for comparison (Ionov et al., 1994; Mattey et al., 1994a; Chazot et al., 1997; Zhang et al., 2000).

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Figure 9: Redox conditions. Details about calculations are given in text, and values aredocumented in Table S1.

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Figure 10: Partitioning (K_D) of trace elements between Cpx and Grt, and between Cpx and Opx. Blue dots represent results of $K_{D(Cpx-Grt)}$ calculations accounting for mineral compositions using the method of Harte and Kirkley (1997). Also shown are data of Johnson (1994), Viljoen et al. (2005), Huang et al. (2007), and Raffone et al. (2009) for comparison.

1222

1223 Figure 11: Whole rock and Ni composition diagrams for studied pyroxenites, and various 1224 lithologies that compose oceanic crust (MORBs = 'volcanic', and the deep plutonic crust 1225 'plutonic'). In all the diagrams, it is clear that the studied pyroxenites differ from oceanic crust lithologies. Also the Ni covariation with MgO highlights the compatible behavior of Ni during 1226 1227 MORB differentiation (Ni decreasing with MgO), and shows that the studied pyroxenites 1228 contains higher amounts of Ni than any MORBs or even of the cumulative rocks from oceanic 1229 crust. Oceanic crust lithologies are presented using density graphs with MORB database 1230 provided by M. O'Hara (n=277), and plutonic rocks database taken from IODP Site U1309 1231 (Godard et al., 2009; n=173).

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1233 Figure 12: P-T-t paths for the studied pyroxenites. Phase diagram for the ultramafic system is 1234 constructed after Gasparik (2014) and references therein. With increasing pressure, 1235 encountered domains are: the *plagioclase lherzolites* facies (A); the *Seiland* (B), and *Ariegites* 1236 (C) sub-facies that correspond to sub-divisions of the *spinel lherzolites* facies (B+C); and the 1237 garnet lherzolite facies (D). Reactions separating different facies when pressure increases are (1): Pl+Ol⇔Cpx+Opx+Spl between A and B (e.g., Takazawa et al., 1996); (2): 1238 1239 $Opx+Spl+Pl \Leftrightarrow Cpx+Grt$ between B and С (e.g., O'Hara, 1967); and (3): 1240 Cpx+Opx+Spl⇔Ol+Grt between C and D (e.g., Johnson et al., 1990). The black line (4) is the
1241 "sapphirine reaction", namely Opx+Pl+Spl⇔Spr+Grt; it occurs only at high temperature and
1242 at slightly higher pressure than reaction (2). S is solidus. The TAK-4 path needs further
1243 constraint, the early P drop is poorly documented and a pure cooling stage is possible for this
1244 sample (dotted line).

1245

1246 Figure 13: Schematic evolution of the petrographic assemblage (a), and qualitative evolution 1247 of the major element (b), and trace element (c) whole rock compositions of the studied mantle 1248 cumulates. The crystallization stage corresponds to the Stage 2 in Figure 14, the 1249 metamorphism stage corresponds to the Stage 3 in Figure 14, and the metasomatism 1250 corresponds to the Stage 3' in Figure 14. In a) the initial cumulative assemblage (or magmatic assemblage from stage 2, marked 'I' in caption), is recrystallized during the subsolidus 1251 1252 evolution (P-T-t variations) to a re-equilibrated metamorphic assemblage (marked 'II' in 1253 caption; Stage 3). Numbered circles represent the possible sampling of the cumulate by a 1254 given xenolith (theoretical samples 1-5). Xenolith '1' only samples Grt, xenolith 2 only 1255 samples Cpx, xenolith 3 only samples Opx, xenolith 4 samples a polymineralic assemblage of 1256 Cpx-Opx-Grt, xenolith 5 samples a polymineralic assemblage of Cpx-Grt. Similar sampling 1257 areas are highlighted at Stage 3 when those initial cumulative assemblages have recrystallized 1258 to metamorphic assemblages. b) and c) highlight that no modification of the whole rock 1259 composition is expected during the subsolidus evolution. Only metasomatism (Stage 3') can 1260 modify mainly the geochemical composition (enrichment in LREE and some other elements, 1261 see Section 8.3 and Table 1). In our theoretical evolution all the samples, but xenolith 3, have suffered metasomatic interactions. REE elements evolution is only qualitative, and based on 1262 1263 minerals' Kds, and on the consideration that metasomatism is associated to LREE enrichment.

1264

1265 Figure 14: General schematic evolutionary model for the studied pyroxenites. At stage 1, an 1266 orogen is formed. At stage 2, the exhumation triggers a sub-isotherm decompression in the 1267 mantle, resulting in partial melting; the formed melt percolates the mantle section, and 1268 crystallizes the studied pyroxenites. After stage 2, studied pyroxenites are crystallized and 1269 their subsequent P-T-t evolution can be followed on Figure 12; their petrographical and 1270 geochemical evolution can be followed on Figure 13. Between stage 2 and 3, the exhumation 1271 is continuous until a thermal re-equilibration. After stage 3 the lithosphere is in a thermal 1272 classical regime, and contains heterogeneities (i.e., the studied pyroxenites), at that stage 1273 mantle domains can suffer metasomatism reactions (=stage 3'). Stage 4: the subsequent 1274 lithospheric evolution (thermal anomaly or rifting initiation), and with modified physical 1275 properties. In particular, the presence of pyroxenite cumulates lowers the global solidus. Compared to lherzolitic mantle regions ('un-refertilized lherzolite mantle'), partial melts will 1276 1277 therefore occur earlier (at lower temperature), and melt production will be higher for a given 1278 temperature in such refertilized areas ('refertilized mantle').

1279

Table 1: Sample list, region, locality, rock type, modal composition, primary magmatic paragenesis, primary magmatic facies, subsolidus paragenesis and features, facies transitions, metasomatic paragenesis or features, late features and alteration phases, microtextures, average grain size, and estimated temperatures for the studied pyroxenites. Modal proportions are given in %, *t* indicates amounts as traces. *FMC* stands for French Massif Central.

1285

Table 2: Major element composition of minerals (in wt %). Same abbreviations as in Table 1. *n* is the number of analyses that is averaged. In the *comment* column, *Opx 1* and *Pl 1* indicate
minerals observed as inclusions in Grt; *reactional* indicates minerals observed in the finegrained Opx-Pl-Spl assemblages in FMC samples (Figure 2d). Data for *Le Pouget* sample are

1290	from Fabriès et al. (1987). %Ph is the An content for Pl, the #Cr for Spl, and the #Mg for			
1291	other phases (molar basis); %Ph2 is the jadeite content for Px, the pyrope content for Grt, and			
1292	the K-feldspar content for Pl.			
1293				
1294	Table 3: Trace element concentration for minerals (ppm). Same abbreviations as in Table 1. n			
1295	is the number of analyses that is averaged. ' and " on sample names indicates that different			
1296	types of minerals are present. Underlined minerals have been analyzed at the IGG (Pavia)			
1297	others at LMV (Clermont-Ferrand).			
1298				
1299	Table 4: Major (wt %) and trace (ppm) element whole rock compositions. LN-78a and LN-			
1300	78b represent two different domains of sample LN-78, and LN-78-avg the averaged value. ba			
1301	means below detection limit.			
1302				
1303	Table 5: Isotopic composition of mineral separates. Same abbreviations as in Table 1. For			
1304	oxygen isotope composition, some samples have been duplicated. std err is the standard error			
1305	on the measurement.			
1306				

1307 Supplementary material:

1308 Figure S1:

a) Oxygen isotope fractionation between Grt and Cpx. For reference, the Δ^{18} O=0 line is 1309 1310 shown (continuous line). The dashed line corresponds to fractionation at 900°C calculated 1311 using the fractionation factors of Zheng (1993). b) Oxygen isotope fractionation between Opx 1312 and Cpx. For reference, is shown the Δ^{18} O=0 line (continuous line), and the field for 1313 lherzolites from Yemen (Chazot et al., 1997). Cpx and Grt oxygen isotopic compositions are 1314 positively correlated and plot below the line corresponding to $\Delta_{Cpx-Grt} = -0.4\%$, indicating that 1315 these minerals have been equilibrated to T above 900° C, in agreement with T calculated 1316 using their chemical composition (see section 8.1.2.). Three samples from Cameroon (YK-01, 1317 YK-03, and YK-05) have positive $\Delta_{Cpx-Grt}$ values and record slight O-isotope disequilibrium. $\Delta_{Cpx-Opx}$ values are negative for all but two samples; the values are heterogeneous but always 1318 1319 above $\Delta_{Cpx-Opx}$ =-0.8‰.

1320

1321 **Table S1:**

1322 Redox calculations results.

1323

1324 Analytical techniques

This study is based on analyses of whole rocks, and minerals that have been separated under
binocular microscope for their purity after crushing, sieving and washing. In-situ analyses
have been made on polished thin sections (30, 100 or 150 µm thick).

Major element mineral compositions were determined using the Cameca SX100 electron microprobe at the "Laboratoire Magmas et Volcans" (LMV) of Université Blaise-Pascal, Clermont-Ferrand, France. Instrument calibration was performed on natural standards and operating conditions were 15kV accelerating potential, 15nA beam current and 15s counting

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time on peak and background. Backscattered electron (BSE) images and EDX X-ray maps
were obtained on the scanning electron microscope at LMV. The modal compositions of
Cameroon samples have been computed from Al, Mg, Ca, Fe mapping with the MultiSpec
software (©Purdue Research Foundation, Inc.
http://dynamo.ecn.purdue.edu/~biehl/MultiSpec/).

1337 Trace element mineral compositions have been determined using a LA-ICP-MS at LMV and 1338 at the CNR-IGG of Pavia. At Pavia the LA-ICP-MS instrument couples a double focusing 1339 sector field ICP mass spectrometer (Element IR from Thermo-Finnigan) with a Q-switched 1340 Nd:YAG laser source (Quantel Brilliant) operating at 213 nm. The laser was operated at a 1341 repetition rate of 10 Hz, and the spot diameter was varied from 20 to 40 µm with a pulse 1342 energy in the range 0.01-0.03 mJ. Helium was used as the carrier gas and was mixed with Ar 1343 downstream of the ablation cell. Quantification has been done using the NIST SRM 612 glass 1344 as external standard, with 44Ca as internal standard. Precision and accuracy were assessed on 1345 the USGS BCR-2(g) reference glass and are both better than 10% for concentration at ppm 1346 level. At LMV, a Q-switched Nd-YAG laser source operating at 266 nm was coupled with a 1347 PQ2+ ICP-MS from Fisons. The laser was operated at a repetition rate of 10 Hz and the spot 1348 diameter was around 60 µm. Other analytical parameters are identical to those at Pavia 1349 laboratory and the precision and accuracy achieved are at the same level.

Whole rock major and trace element concentrations have been determined by a lithium metaborate fusion method and using an ICP-AES and an ICP-MS, respectively at the Activation Laboratories (Ontario). For sample from "Le Pouget" (FMC), the available sample amount was very small and the whole rock trace element content has been estimated using the mineral trace element compositions, and the modal proportions.

1355 The oxygen isotopic composition of mineral separates was analyzed at the CNR-IGG of Pisa 1356 (Italy), by conventional laser fluorination, reacting the samples under a F_2 gas atmosphere.

1357 For each sample, 1 to 2mg of separated grains were placed under a 25 W Merchantek CO₂ 1358 laser. The produced reaction triggers the oxidation of the different mineral molecules, and 1359 liberates the O₂ following reactions as $2MgO+SiO_2+xF_2 \Leftrightarrow 2MgF_2+SiF_4+2O_2+(x-4)F_2$ (with a 1360 large x in order to achieve a high total yield). The gas mixture was injected in a separation line 1361 in which different cold trap and heated salts (KCl at ~140°C) allow us to trap all the molecules but O₂. The purified O₂ is thereafter injected in a mass spectrometer (Finnigan 1362 MAT Delta plus XP) to analyze the different isotopes. A laboratory inner reference gas is 1363 1364 jointly analyzed to compare the analyzed sample with the V-SMOW international standard.

1365 The
$$\delta^{18}$$
O value is then calculated (with δ^{18} O% = $\frac{\overset{e}{\hat{e}}}{\overset{e}{\hat{e}}} \frac{{}^{18} \text{O}/{}^{16} \text{O}}_{(\text{V-SMOW})} - 1 \overset{u}{\overset{u}{\hat{u}}} + 1 \overset{u}{_{\hat{u}}} +$

average reproducibility of oxygen isotope analyses of silicate minerals is 0.16‰. Standards were analyzed at the beginning and end of each analytical series; these were the in-house laboratory standard QMS quartz (theoretical δ^{18} O value =14.05 ‰), and UWG-2 garnet (theoretical δ^{18} O value=5.8 ‰).

The Sr-Nd radiogenic isotopic analyses have been achieved at LMV using a thermo ionization 1370 1371 mass spectrometer (TIMS) Triton from Thermo-Finnigan. Each analysis has been accomplished on 100 mg of separate Cpx. Sr and Nd were extracted following a 1372 1373 chromatographic extraction method after having effectuated a leaching: 30min in HCl 2.5N, 1374 15min in HF 5%, 30min in HCl 2.5N and 30min in distilled water. Samples were then 1375 evaporated on tungsten filaments and analyzed using the double filament method with the 1376 TIMS Finnigan Triton. Standards used for this study are BIR-1 0.703136±4 and NBS987 1377 0.710279±4 for Sr and BIR-1 0.513111±4 and AMES 0.511960±3 for Nd.





Initiation of thermal anomaly

- + Upper crust
- 🔶 Lower crust
- × Pyroxenites mantle source
- × Mantle partial melting
- Melt migration
 - Pyroxenites crystallization
- Studied pyroxenites



France et al., Figure 1















France et al., Fig. 2; continuous

Um

G

mr





France et al., Fig. 4





France et al., Fig. 6







France et al., Figure 9



France et al., Fig. 10





France et al., Fig. 12





Stage 4 Post orogenic initiation of thermal anomaly or mantle plume



+	Up	per	crust

- ✓ Lower crust
- × Pyroxenites mantle source
- × Mantle partial melting
- Melt migration
- Pyroxenites crystallization
- Studied pyroxenites


France et al., Fig. S1